

# PATENT ABSTRACTS OF JAPAN

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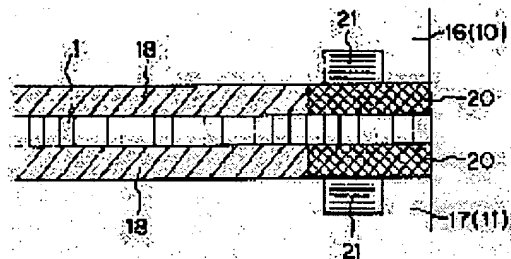
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## (54) SEALING METHOD FOR SOLID POLYMERIC FUEL CELL

### (57)Abstract:

**PURPOSE:** To provide a sealing method ensuring freedom from damages to a polymeric electrolyte film, and ensuring safety by densifying the peripheral section of an electrode for a solid polymeric fuel cell and applying a gas seal to the densified section.

**CONSTITUTION:** The peripheral sections of electrodes 18 and 18 for a solid polymeric fuel cell are densified, and the densified sections 20 and 20 are used for gas sealing with a packing 21, in conjunction with frames 10 and 11, and separators 16 and 17. This densifying method is not limited in particular. An example of the method, however, is that the peripheral sections of the electrodes 18 and 18 are impregnated with a solution of a densifying agent formed out of an adhesive or the like, and a solvent is removed from the solution whenever necessary. Also, the densifying agent to be used is preferably a polytetrafluoroethylene resin dispersed agent. This densifying agent is in particular effective for the gas diffusion electrode where a catalyst layer made of catalyst particulates, an electrolyte and polytetrafluoroethylene is on water repellent carbon paper.



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CLAIMS

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[Claim(s)]

[Claim 1] The seal approach of the polymer electrolyte fuel cell which carries out eburnation of the periphery section of the electrode for polymer electrolyte fuel cells, and is characterized by performing a gas seal by this eburnation part.

[Claim 2] The seal approach of a polymer electrolyte fuel cell according to claim 1 of performing the above-mentioned eburnation by carrying out impregnation of the eburnation agent to the periphery of the electrode for polymer electrolyte fuel cells.

[Claim 3] The seal approach of the polymer electrolyte fuel cell according to claim 1 or 2 which is the electrode which made the catalyst bed to which the above-mentioned electrode consists of a catalyst particle, an electrolyte, and polytetrafluoroethylene on \*\*\*\*\* carbon paper support.

[Claim 4] The seal approach of the polymer electrolyte fuel cell according to claim 1, 2, or 3 which uses the resin of a polytetrafluoroethylene system as an eburnation agent.

[Claim 5] The seal approach of a polymer electrolyte fuel cell according to claim 1, 2, 3, or 4 that the solid-state polyelectrolyte film of a polymer electrolyte fuel cell is film of perfluorocarbon sulfonic acid system resin.

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DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[Industrial Application] Without more specifically damaging the solid-state polyelectrolyte film about the seal approach of a polymer electrolyte fuel cell, this invention makes the gas seal of a polymer electrolyte fuel cell easily and reliable, and relates to the seal approach of the polymer electrolyte fuel cell which can raise the safety of a polymer electrolyte fuel cell effectively.

[0002]

[Description of the Prior Art] Although a polymer electrolyte fuel cell has the description at the point which an ion conductor, i.e., an electrolyte, is a solid-state, and is a macromolecule As the solid-state polyelectrolyte, film, such as ion exchange resin, is specifically used. this polyelectrolyte film -- inserting -- the two electrodes of a negative electrode (anode) and a positive electrode (cathode) -- arranging -- for example, a negative-electrode side -- the hydrogen gas as a fuel -- moreover, the electrical and electric equipment is generated by supplying oxygen or air to a positive-electrode side, and making electrochemical reaction cause.

[0003] Although there is a thing of various modes in this equipment until now, drawing 1 is a schematic diagram for explaining one mode of this polymer electrolyte fuel cell. As for the polyelectrolyte film and 2, one is [ a cathode electrode (positive electrode) and 3 ] anode electrodes (negative electrode) among drawing 1 , and the polyelectrolyte film 1 is arranged in contact with between these positive/negative two electrodes 2 that face, and 3. Moreover, 4 is a cathode electrode side charge collector, 5 is an anode electrode side charge collector, and it is contacted by the electrodes 2 and 3 of positive/negative, respectively.

[0004] Among these, the slot for oxygen or air supply is established in the electrode 2 side of the cathode electrode side charge collector 4, the slot for [ it is the same and ] hydrogen supply in the electrode 3 side of the anode electrode side charge collector 5 is prepared, the slot of the positive-electrode side charge collector 4 is open for free passage in oxygen or the air supply tubing 6, and the slot of the negative-electrode side charge collector 5 is open for free passage to the hydrogen supply pipe 7. Moreover, it is the cathode terminal assembly with which 8 was prepared in contact with the positive-electrode side charge collector 4, and the anode terminal assembly with which 9 was prepared in contact with the negative-electrode side charge collector 5, and power is taken out through these during actuation of a cell. Further 10 is an up frame (up frame), and 11 is a lower frame (lower frame), and the cell proper from the polyelectrolyte film 1 to the cathode terminal assembly 8 and the anode terminal assembly 9 is covered with these vertical both the frames 10 and 11, and it is fixed.

[0005] Between these vertical both the frames 10 and 11, the periphery section of the cell proper from the polyelectrolyte film 1 to the cathode terminal assembly 8 and the anode terminal assembly 9 is enclosed, packing (gasket) 12 is formed, by this, it fixes densely, the seal of the periphery section of the cell proper is carried out, and the gas seal is especially carried out to the polyelectrolyte film 1 and the positive/negative two electrodes 2 and 3. In addition, 13 and 14 are cooling water supply pipes among drawing 1 , and these are open for free passage into the slot (close path) established in the inner surface

of the up frame 10 and the lower frame 11, respectively, and are cooled from the tooth back of the cathode terminal assembly 8, and the tooth back of the anode terminal assembly 9.

[0006] Although the above is the case that a cell proper is single, accumulating and constituting these two or more cells proper is also performed. In this case, although it is necessary to make a separator intervene between each two or more cells proper, and to establish the slot for cooling water etc. also in this, it is fundamentally [ including enclosing the periphery section of a cell proper, preparing packing, fixing densely, carrying out the seal of the periphery section of that cell proper, and carrying out a gas seal to the polyelectrolyte film 1 and the positive/negative two electrodes 2 and 3 etc. ] the same as the case of the cell proper of the above-mentioned single. In this case, bolting of packing 12 grade is performed also through a separator.

[0007] In this case, as the method of that seal, the technique of making \*\* O ring to which packing is made to be placed between and stuck placed between the perimeters of the polyelectrolyte film, and making it stick etc. by this is used and proposed until now as \*\* above-mentioned. Drawing 2 shows the method of the seal by this \*\* O ring. electrodes 2 and 3 arrange to the vertical side of an electrolyte membrane 1 as a graphic display -- having -- the upper and lower sides to vertical both the frames 10 and 11 -- moreover -- the case where two or more cells proper are accumulated and constituted -- vertical both the frames 10 and 11 -- in addition, a seal is carried out by making O ring 15 arranged between the periphery sections of these and an electrolyte membrane 1 intervene, and sticking it with separators 16 and 17. In addition, among drawing 2, what are indicated to be a sign "16 (10)" and "17 (11)" means that it will be equivalent to the up frame 10 or the lower frame 11, when separators 16 and 17 serve as the topmost part or the bottom.

[0008] However, by the technique of these \*\*s and \*\*, it is contacted by whether packing or an O ring is direct on the polyelectrolyte film 1, in order to ensure that adhesion, each needs to press these packing or an O ring strongly, and bolting beyond the need becomes for this reason, it not only damages the polyelectrolyte film itself, but performed to the above-mentioned whole cell proper. Moreover, an electrolyte membrane usually has the property expanded and contracted by temperature or the existence of humidification, and since a burden tends to be placed on the seal section by this, even if it takes which the above conventional seal technique, it is necessary to also consider this point.

[0009]

[Problem(s) to be Solved by the Invention] Then, it hits this invention fixing and carrying out the seal of the cell proper, and carrying out a gas seal to the polyelectrolyte film and positive/negative two electrodes. In contact with [ as / in technique, such as the aforementioned \*\* and \*\* ] whether to be direct, the seal of packing or the O ring is not carried out at the periphery section of the polyelectrolyte film. It aims at not damaging the polyelectrolyte film and offering the seal method safety is high by being made to carry out a seal in the periphery section of the two electrodes moreover arranged on the underside.

[0010]

[Means for Solving the Problem] This invention offers the seal approach of the polymer electrolyte fuel cell which carries out eburation of the periphery section of the electrode for polymer electrolyte fuel cells, and is characterized by performing a gas seal by this eburation part.

[0011] Usually the electrode for polymer electrolyte fuel cells is applied in a sheet-like (shape of film) form. As the sheet-izing \*\* Make the electrode component containing a catalyst particle adhere to whether it is direct to the solid-state poly membrane as an electrolyte. \*\* Although carried out in the various modes of making the suspension containing \*\* catalyst particle sheet-ized with rolling etc. by using the electrode component containing a catalyst particle as a kneading object adhere on the \*\*\*\*\* carbon paper as a base material sheet (for it to become a gaseous diffusion layer in an electrode) etc. This invention is applicable also to the electrode obtained in which these modes.

[0012] this invention -- the voice of the above-mentioned \*\* -- especially when taking the mode which makes the suspension of the catalyst component (it becomes a catalyst bed in an electrode) which consists of \*\*\* catalyst particle, an electrolyte, and polytetrafluoroethylene adhere on the \*\*\*\*\* carbon paper as a base material sheet (for it to become a gaseous diffusion layer in an electrode), it is

advantageous also in inside [ like ], and it is desirable that it is the thing of a polytetrafluoroethylene system as a water repellent agent of that paper in this case. A polytetrafluoroethylene system is the semantics containing an others and tetrafluoroethylene-hexafluoropropylene copolymer, its other copolymers, etc. here. [ polytetrafluoroethylene ]

[0013] Moreover, if it is the technique of the ability to carry out eburnation of the periphery section of an electrode as the method of the above-mentioned eburnation in this invention, especially definition can carry out impregnation of the solution of an eburnation agent which there is not, for example, consists of adhesives etc. to the periphery section, and can be performed by removing a solvent from the solution if needed. Although it can be used if it is a \*\*\*\*\* thing as this eburnation agent to carry out eburnation of the electrode periphery section, it is desirable that it is the eburnation agent which has the thermal resistance which it is not necessary to degrade especially an electrode and the polyelectrolyte film according to chemical action etc., and can be borne with the operating temperature of a cell, and acid resistance.

[0014] Although the dispersion of a polytetrafluoroethylene system (an others and tetrafluoroethylene-hexafluoropropylene copolymer, its other copolymers, etc. are included) can be used preferably as this eburnation agent, it is effective to the gas diffusion electrode which made the catalyst bed adhere on the above-mentioned \*\*\*\*\* carbon paper, and especially this eburnation agent is effective especially when using the film of the below-mentioned perfluorocarbon sulfonic acid resin system as polyelectrolyte film. [ polytetrafluoroethylene ]

[0015] In the electrode for polymer electrolyte fuel cells of the format that this invention person uses a catalyst particle, a polyelectrolyte, and the mixture of polytetrafluoroethylene (in addition) The electrode of this format is manufactured by any mode of the above-mentioned \*\* - \*\*. The approach of improving that conductivity etc. by coating with the whole surface of the surface layer of this electrode with solid-state polyelectrolyte solutions, such as for example, a perfluorocarbon-sulfonic-acid resin system, is developed previously. although proposed (Japanese Patent Application No. No. 297281 [ five to ]) -- this invention -- for example, it is effectively applicable also about the fuel cell using the electrode obtained by doing in this way.

[0016] Drawing 3 - drawing 4 are the schematic diagrams for explaining the seal method of this invention. Among these, drawing 3 uses \*\*\*\*\* carbon paper as a gaseous diffusion layer as an example, the electrode sheet (gas diffusion electrode) 18 of the format of having made the one side depositing and supporting a catalyst bed is shown, and the periphery part which processed 19 with \*\*\*\*\* by water-repellent \*\*\*\*\* of \*\*\*\*\* carbon paper among drawing 3 , processed 20 by the eburnation agent, and carried out eburnation is shown. Moreover, drawing 4 shows the condition of having carried out the seal of such processing using the electrode sheet 18 which obtained by carrying out, among drawing 4 , although 21 are up-and-down packing, in this invention, it cannot restrict but other means, such as an O ring, can be applied with packing.

[0017] Although it is made for an electrolyte membrane 1 to contact the electrode sheet 18 as drawing 4 in the vertical both sides, the whole surface The film surface of the electrode sheet 18 has the same area as the front face of an electrolyte membrane. By this for this reason, packing 21 It does not touch to an electrolyte membrane side, but is arranged in contact with the periphery aspect [namely, the part 20 which processed by the eburnation agent and carried out eburnation with \*\*\*\*\*] of the electrode sheet 18. This with vertical both the frames 10 and 11 from the upper and lower sides Moreover, when it accumulates and constitutes two or more cells proper, in addition to vertical both the frames 10 and 11, a seal is carried out by making it stick with separators 16 and 17.

[0018] In this invention, by making packing (or O ring etc.) contact this eburnation part, eburnation of the periphery section of an electrode can be carried out in this way, a seal is carried out, breakage on an electrolyte membrane can be prevented, it can be a total, and the safety of a cell can be raised by this. In addition, although the film surface of an electrode sheet has the same area as the front face of an electrolyte membrane in the mode shown in drawing 4 , it is necessary to be the limit where the film surface product of this electrode sheet makes packing (or O ring etc.) contact the eburnation part of the periphery section of that electrode, and the predetermined seal object can be attained, and the surface

area of an electrolyte membrane does not necessarily need to be the same area.

[0019] As an electrolyte membrane for polymer electrolyte fuel cells It improves serially from the condensation synthetic membrane of \*\* phenolsulfonic acid of the beginning, and formaldehyde. The film sulfonated after carrying out the crosslink of the polystyrene film and \*\* styrene-divinylbenzene which it was improved and were sulfonated on \*\* partial target until now to the matrix of fluorocarbon, \*\* The film which does not include alphaC-H coupling by the film of \*\*, the polymerization film of \*\* trifluoro styrene sulfonic acid, the film which graft-ized trifluoro ethylene to \*\* fluorocarbon matrix, \*\* perfluorocarbon sulfonic acid resin film, etc. are proposed.

[0020] Chemically stable in itself the seal approach concerning this invention -- the polyelectrolyte film of these instantiation -- not restricting -- the class of polyelectrolyte film -- how -- not asking -- application -- possible -- moreover -- the material of packing -- a fluororubber and others -- If it is equipped with many predetermined properties as packing that fluids, such as hydrogen and air, do not permeate etc., without deteriorating the ingredient in contact with this, the same is said of that construction material in the case of being able to use all and using this point O ring etc.

[0021] moreover, the resin film (NAFION, trade name) of \*\* perfluorocarbon sulfonic acid system among the polyelectrolyte film of said instantiation -- those outstanding electrical characteristics -- adding -- current, since it is physical very [ chemically or ] stable and a machine target is also large, [ionic conductivity  $5 \times 10^{-2}$  S-cm<sup>-1</sup> (a damp or wet condition, 25 degrees C)] and -- this resin film is used as main. This film is used as film with a thickness of about 50-200 micrometers, and although the electric resistance per unit area is so small that it cannot cause [ main ] internal resistance of a cell by about 0.1-0.5ohms, this thickness In using this film as polyelectrolyte film the solution of the polyelectrolyte film desirable and same as the adhesives as this -- [ -- for example, if the film of Nafion117 film (trademark) is used as polyelectrolyte film, Nafion solution (product [ made from Aldrich Chemical ], trademark)] will be used.

[0022] Hereafter, although the example of this invention is explained, of course, it is not that by which this invention is limited to this example. \*\* After making a carbon paper side with a porosity [ 2 and 80% of porosity ] of with a surface area of 10x10cm, and a thickness of 0.4mm first carry out impregnation of the dispersion of the neo chlorofluocarbon (a tetrafluoroethylene-hexafluoropropylene copolymer, the Daikin Industries, LTD. make, trademark) diluted to 12.5%, the carbon paper which performed heat treatment at the temperature of 380 degrees C for 3 hours, and \*\*\*\*\* (ed) by neo chlorofluocarbon was obtained. About the quantitative rate, it considered as 80 % of the weight of carbon paper, and 20 % of the weight of neo chlorofluocarbon here.

[0023] \*\* After carrying out impregnation of Pori Flon's (polytetrafluoroethylene, DANKIN industrial company make, trademark) dispersion to the periphery of the \*\*\*\*\* carbon paper obtained by \*\* 60% of the weight subsequently, at the temperature of 380 degrees C, heat treatment was performed for 3 hours and the carbon paper which carried out eburation of the periphery by Pori Flon was obtained. \*\* In addition, the eburation of the carbon paper obtained in the activity of \*\* is inadequate, and when gas carries out a leak, eburation can fully be carried out by repeating the same actuation.

[0024] On this periphery eburation carbon paper side, succeedingly \*\* (Remove a surrounding eburation part), By making the suspension which added the dispersion of polytetrafluoroethylene to the catalyst particle which makes it come to the carbon particle with which it comes to coat with the alcoholic solution of perfluorocarbon-sulfonic-acid resin to support 50 % of the weight of platinum deposit The electrode sheet as shown in drawing 3 was obtained by making a catalyst particle and a water repellent agent (polytetrafluoroethylene) deposit on the eburation carbon paper, and carrying out impregnation of the polyelectrolyte solution further. \*\* Nafion-117 film (the solid-state polyelectrolyte film, the product made from Du Pont, trade name) was further inserted between the electrode sheets of two sheets obtained above, and the body of a fuel cell was acquired.

[0025] Subsequently, the charge collector, the terminal assembly, etc. were stuck on the above-mentioned body of a fuel cell like drawing 1 with the conventional method, and the gate of hydrogen and oxygen etc. was installed, it set as a polymer electrolyte fuel cell (about the seal part, it carried out as drawing 4 ), and change of the engine performance as the electrode characteristic and a cell was